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COMPLETE SPECIFICATION

Stabilized Homopolymers, Copolymers and Derivatives of Alkylene Oxides

We, FARBWERKE HOECHST AKTIENGESELLSCHAFT, vormals Meisten Lucius & Brüning, a body corporate recognised under German Law, of 6230 Frankfurt (M)-Hoechst, Germany, do hereby declare the invention, for which we pray that a patent may be granted to us, and the method by which it is to be performed, to be particularly described in and by the following statement:—

This invention relates to the stabilization of homo- and co-polymers of alkylene oxides and derivatives thereof.

Homo- and copolymers of alkylene oxides, as well as their addition products with substances which contain active hydrogen, are subject to oxidative decomposition in the presence of light and oxygen which decomposition generally increases when the temperature is raised. The decomposition, which constantly increases, is shown, for example by a decrease in the viscosity of aqueous solutions of the polymers, an increase in their acid number and by an increase of their aldehyde content.

High stability against oxidative decomposition is necessary when using such polymers of alkylene oxides, for example, as lubricants, as components for hydraulic liquids, as operating liquids for hydrodynamic power transmissions, as heat transmission media, as thickening agents and binding agents, as carriers in the fields of pharmacy and cosmetics, for chemico-technical preparations, as dyestuff media, as textile auxiliaries and as viscose-modifiers. High stability can often only be achieved by using an appropriate stabilizer.

Stabilization against oxidative decomposition is also important in the maintenance of constant physical properties, for example, the viscosity and the lubricity, of technical operating liquids. Small amounts of aldehydes or

acids, which are formed during oxidative decomposition, may have extremely disturbing effects and lead to corrosion or attack and transform sensitive substances, for example albumen, odoriferous substances or therapeutical substances, and influence the pH-value of their aqueous solutions. Polyalkylene oxides, the molecular weight of which lies above about 50,000 are especially endangered by oxidative decomposition, and must be stabilized before being used.

Hitherto proposed stabilizers are, for example, aromatic amines and phenols, but their action is often insufficient to prevent decomposition successfully. The requirements for such stabilizers regarding, for example, their solubility in the media to be stabilized and the need to prevent discolouration and other undesired attendant phenomena, vary considerably according to the desired application and quite special types of stabilizers having rather complicated chemical structures have gradually been developed. Multi-component mixtures have also been proposed (German Patents 1,210,179, 1,210,558, 1,210,560, 1,211,389, 1,218,729, 1,218,730, 1,210,558).

The present invention is based on the observation that stabilization of homo- and copolymers of alkylene oxides and reaction products of alkylene oxides with substances containing active hydrogen can be relatively simply effected by adding to them or their solutions minor amounts of powdered manganese metal or of a manganese-containing compound.

The amount of powdered manganese or manganese compound used is advantageously in the range of from 0.0005 to 5%, preferably 0.005 to 0.5%; based on the weight of the polymer to be stabilized.

Mixtures of manganese metal and one or

more manganese-containing compounds or of two or more manganese-containing compounds may also be used as a stabilizer.

Amongst the suitable manganese-containing compounds there may especially be mentioned manganese (II)-salts of inorganic and organic acids, for example, manganese-(II)-chloride, manganese-(II)-sulphate, manganese-(II)-nitrate, manganese-(II)-acetate and manganese-(II)-resinate. (IV)-valent manganese compounds, for example manganese dioxide, in finely distributed form also have a well-defined stabilizing action against oxidative degradation. Potassium permanganate also has a well-defined stabilising action.

Manganese powder is advantageously used in finely distributed form having a grain size of less than 150μ , preferably of less than 40μ . Commercial manganese powders may be used, which may also contain impurities, for example, carbon or iron.

The stabilising action of the manganese compounds or the metallic manganese is not reduced even in strong acid medium and is not reduced even in the presence of the hitherto proposed antioxidants. It is therefore possible according to the present invention to stabilise aqueous mineral acid solutions of homo- and copolymers and derivatives of ethylene oxides.

The addition of manganese compound or powder is carried out by dissolving or suspending it in the medium which is to be stabilized. Some manganese compounds, for example, manganese-(II)-chloride, manganese-(II)-nitrate or manganese-(II)-resinate have a relatively good solubility in polyethylene-glycols dissolution being facilitated by heating. $MnCl_2 \cdot 4H_2O$ is extremely soluble in polyethylene glycols. If for example 3 parts by weight of this compound are heated to $117^\circ C$ together with only 2 parts by weight of liquid polyethylene glycol having a molecular weight of about 400 there is obtained a weakly yellowish solution, which, after cooling, is highly viscous and clear. Thus, it is possible to prepare high-percentage concentrates of such manganese compounds in polyethylene glycols, which then can be added to the product to be stabilized in the corresponding amounts. In polyethylene glycols of higher molecular weight which are solid at room temperature the solution or dispersion is carried out after melting. Other manganese compounds for example, the sulphate, which are insoluble in polyethylene glycols may nevertheless advantageously be used for stabilizing their aqueous solutions against oxidative decomposition.

The solubility of the manganese compounds in other polyalkylene-glycols (e.g. co-polymers of ethylene oxide and propylene oxide) is generally lower. The addition of the manganese compounds can be carried out in dissolved form with the aid of an appropriate solvent. Thus, for example, stabilization of poly-propylene-glycol having a molecular weight of 2000, in which manganese-(II)-chloride is practically insoluble, can be carried out by dissolving the chloride in dipropylene glycol and then adding this solution to the batch. In addition, the good solubility of many manganese compounds in water allows the stabilization of aqueous solutions of polyalkylene glycols and their derivatives.

These aqueous solutions are important in many practical applications.

As examples of the homo- and copolymers of alkylene oxides and reaction products of alkylene oxides and substances containing active hydrogen which may be stabilized by the addition of manganese compounds of manganese powders there are mentioned polyalkylene oxides, for example, polyethylene glycol and polypropylene glycol, copolymers of ethylene oxide and propylene oxide and the reaction products of ethylene oxide or propylene oxide with, for example, alcohols, for example stearyl or oleyl alcohol, or carboxylic acids, for example, stearic acid or oleic acid. The stabilizing agents which are used according to the process of the present invention have a better stabilizing action than the hitherto proposed stabilizing agents. A further advantage of the manganese compounds or manganese powders used according to the present invention lies in the fact that they are physiologically tolerable.

The following Examples illustrate the invention:

EXAMPLE 1.

0.057% of manganese-(II)-chloride (referred to polyalkylene glycol) are dissolved in a 50% aqueous solution of a polyalkylene oxide, consisting of ethylene oxide and propylene oxide in a molar ratio of 1:3 and having a molecular weight of 11,000 and which is used as starting substance for a difficultly flammable hydraulic liquid. During 30 hours, 6 litres of oxygen per hour are passed through the solution at $95^\circ C$. A corresponding test without addition of manganese-(II)-chloride is made as comparison. The oxidative decomposition as to viscosity, acid number and content of aldehyde is compared.

polyalkylene glycol mol. weight 11000 in- a 50% aqueous solution	viscosity at 25°C	acid number referred to the undilute product	content of aldehydes; calculated as acetaldehyde
Before the heat test	337 cP	0.35	0.017%
after 30 hours heat test without stabilizer	74 cP	37.7	0.129%
after 30 hours heat test w. addition of 0.057 % of $MnCl_2$	313 cP	11.6	0.014%

EXAMPLE 2

A 25% aqueous solution of a polyethylene glycol of an average molecular weight of about 35,000, to which 0.03% of manganese-(II)-

chloride is added, is submitted to a heat test as described in Example 1 and compared with an equally treated polyethylene glycol-solution which is free of manganese.

polyethylene glycol mol. weight 35000 in 25% aqueous solution	viscosity at 50°C	acid number referred to the pure polyethylene glycol	content of formaldehyde in %	total content of aldehydes calculated as acetaldehyde in %	content of peroxides calculated as H_2O_2 in %
Before the heat test	400 cP	0.15	0.004	0.016	0.0009
after 30 hours heat test without stabilizer	18 cP	56.5	0.029	0.27	0.006
after 30 hours heat test with 0.03% of $MnCl_2$	360 cP	0.8	0.002	0.009	0.0005

EXAMPLE 3

0.0023% of manganese-(II)-chloride (referred to the weight of the solution) are added to a 1% aqueous solution of polyethylene glycol having a molecular weight of about 500,000 and heated to 80° C. for 6 hours. Air or additional oxygen is not bubbled through, since the polyethylene oxides of such high molecular weight which are appropriate for textile

auxiliaries as well as thickening and binding agents for pharmaceutical, cosmetic and chemo-technical products, when heated are increasingly submitted to decomposition. They are distinguished by a high viscosity of their aqueous solutions, which rapidly decreases with the decomposition of the product. The comparison with the similar polyethylene oxide without stabilizer gives the following results:

polyethylene oxide molecular weight 500 000 in 1% aqueous solution	viscosity of the solution at 25°C
Before the heat test	165 cP
after 6 hours heating to 80°C without addition of $MnCl_2$	8 cP
after 6 hours heating to 80°C with addition of 0.0023% $MnCl_2$ referred to the weight of the prepared solution	151 cP

EXAMPLE 4.
20% aqueous solutions of the poly addition products of ethylene oxide to stearic acid or
5 stearyl alcohol are submitted to the heat test

as described in Example 1 by simultaneously passing through oxygen and compared with the corresponding solution without addition of manganese.

tested product in 20% aqueous solution	acid number referred to the undiluted product	content of formaldehyde %	total content of aldehydes, calculated as acet-aldehyde %	content of peroxides calculated as H_2O_2 %	appearance
stearic acid +25 mols of ethylene oxide (EO) before the heat test	0.6	0.0006	0.005	0.0006	water clear solution
stearic acid +25 mols EO without stabilizer after 30 hours heat test	106.0	0.0106	0.2	0.0074	milky white dispersion
stearic acid +25 mols EO with 0.025% of $MnCl_2$ after 30 hours heat test	0.6	0.001	0.006	0.0006	water-clear solution
stearyl alcohol +25 mols EO before the heat test	0.5	0.0007	0.007	0.004	water-clear solution
stearyl alcohol +25 mols EO without stabilizer after 30 hours heat test	57.0	0.0134	0.1	0.007	milky white dispersion
stearyl alcohol +25 moles EO with 0.025% of $MnCl_2$ after 30 hours heat test	2.8	0.002	0.001	0.0009	water-clear liquid

EXAMPLE 5.

A mixture of 96% of polypropylene glycol of an average molecular weight of 2000 and 4% of dipropylene glycol is submitted to the heat test as described in Example 1 by simultaneously passing through oxygen and com-

pared with the corresponding non-stabilized product. Since most manganese compounds are insufficiently soluble in polypropylene glycols for the process of the present invention dipropylene glycol was used as solvent.

96% of polypropyleneglycol 2000 +4 % of dipropylene glycol	viscosity at 25°C	acid number
Before the heat test	318 cP	0.1
after 30 hours heat test without stabilizer	161 cP	49.8
after 30 hours heat test with addition of 0.03% of $MnCl_2$	298 cP	2.2

EXAMPLE 6.

Pure polyethylene glycol having an average molecular weight of 400 shows under the conditions of a heat test mentioned in Example 1, phenomena of decomposition, which can be

read off from the acid number and aldehyde content. The following survey shows that the addition of 0.114% of manganese chloride largely prevents the oxidative decomposition.

polyethylene glycol 400	acid number	aldehyde content, calculated as acetaldehyde	formaldehyde	peroxide content calculated as H_2O_2
Before the heat test	0.1	0.028%	0.0011 %	0.0017 %
without addition of stabilizers after 30 hours heat test	12.0	0.37%	0.12 %	0.02 %
with 0.114 % of $MnCl_2$ after 30 hours of heat test	0.1	0.016%	0.0035 %	0.0005 %

EXAMPLE 7.

Dilute aqueous solutions of polyethylene glycol having a molecular weight of about 1500 of different concentrations are heated under reflux to 80° C during a period of 140-430 hours in the presence of different stabilizers. Tests 10-16 contain additions of 5% sulfuric acid, 11% of sodium sulfate and 7% zinc sulfate as they are usual in acid precipitating baths in the viscose industry. In the following Table they are called "acid bath". The activity of known stabilizers is used for comparison. After the test the amount of formaldehyde which has

formed as characteristic is determined for the decomposition of the polyethylene glycol.

In tests 8 and 9 the content of undecomposed polyethylene glycol which remained was tested. In test 8 only half of the originally used amount was to be found whereas in test 9 the originally used polyethylene glycol was to be found in practically the same amount. The determinations have been carried out according to K. Bürger "Trace analysis and quantitative determination of polyethylene glycols" (Zeitschrift Analyt. Chem. 196-259 (1963)).

TABLE TO EXAMPLE 7

test No.	aqueous solutions of glycol (PEG) 1500 PEG-content	polyethylene additions	duration of heating, hours	formaldehyde ppm
1	1%	none	232	78
2	1%	MnCl ₂ /10 ppm Mn	232	<1
3	1%	MnCl ₂ /2.5 ppm Mn	216	<1
4	1%	MnSO ₄ /10 ppm Mn	216	<1
5	1%	Mn(NO ₃) ₂ /10 ppm Mn	216	<1
6	1%	Mn-acetate/10 ppm Mn	216	<1
7	1%	Mn-citrate-10 ppm Mn	216	<1
8	0.1%	none	232	24
9	0.1%	MnCl ₂ /10 ppm Mn	232	<1
10	0.1%	acid bath without stabilizer	216	24
11	0.1%	acid bath MnSO ₄ / 10 ppm Mn	216	<1
12	0.1%	acid bath MnCl ₂ /5 ppm Mn	430	3
13	0.1%	acid bath 10 ppm 10 ppm phenothiazine	430	33
14	0.1%	acid bath 10 ppm propylgallate	142	21
15	0.1%	acid bath 10 ppm phenyl-β-naphthylamine	142	17
16	0.1%	acid bath 10 ppm tert- butyl-hydroxyanisole	142	18

EXAMPLE 8.

5 A 25% aqueous solution of a polyethylene glycol having a molecular weight of 35000 is heated under reflux to 95° C for 24 hours for comparison with and without stabilizer by simultaneously passing through 6 litres of oxygen per hour and the oxidative decomposition is tested by means of the viscosity. For

this process, besides the manganese-(II)-sulfate, 10 known stabilizers are also used for comparison. Furthermore, the amount used of a stabilizer is varied between 0.004—0.0004% (referred to the polyethylene glycol) used. The excellent stabilizing action of the manganese becomes 15 most clear with only small amounts thereof.

Amount of stabilizer referred to the weight of the PEG 35000	duration of heating	viscosity of the aqueous solution at 25°C	hours	viscosity	hours	viscosity
without stabilizer before the heat test	0	380 cP	—	—	—	—
without stabilizer after the heat test	8	88 „	16	33 cP	24	17 cP
0.004% MnSO_4	8	364 „	16	349 „	24	299 „
0.004% Phenothiazine	8	359 „	16	177 „	24	57 „
0.004% PBN+	8	175 „	16	55 „	24	27 „
0.004% PAN++	8	336 „	16	180 „	24	50 „
0.004% Propylgallate	8	106 „	16	74 „	24	16 „
0.001% MnSO_4	8	348 „	16	167	—	—
0.001% Phenothiazine	8	124 „	16	44	—	—
0.001% PBN+	8	129 „	16	53	—	—
0.001% PAN++	8	169 „	16	76	—	—
0.0004% MnSO_4	8	273 „	—	—	—	—
0.0004% Phenothiazine	8	141 „	—	—	—	—
0.0004% PBN+	8	123 „	—	—	—	—
0.0004% PAN+	8	146 „	—	—	—	—

+PBN= phenyl- β -naphthylamine

+PAN= phenyl- α -naphthylamine

EXAMPLE 9

0.025% of manganese powder having a grain size of $<40\mu$ (referred to the polyalkylene glycol) is added to a 50% aqueous solution of a polyalkylene glycol consisting of ethylene oxide and propylene oxide in a molar ratio of 3:1 and having a molecular weight of about

11000. 6 litres of oxygen per hour are passed through the mixture at 95°C during 30 hours. A corresponding test was carried out without the addition of manganese powder for comparison. The oxidative decomposition as to viscosity, acid number and aldehyde content was compared.

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polyalkylene glycol mol. weight 11000, in 50% aqueous solution	viscosity at 25°C	acid number referred to undilute product	content of aldehydes calculated as acetaldehyde
before the heat test	337 cP	0.35	0.017 %
after 30 hours heat test	74 cP	37.7	0.129 %
after 30 hrs. heat test w. add. of 0.025% of manganese powder	319 cP	0.8	0.01 %

When using manganese powder having a grain size between 80 and 120 μ similar results are obtained.

glycol having an average molecular weight of 35000, to which 0.012% of manganese powder is added is submitted to a heat test as described in Example 1 and compared with a polyethylene glycol solution free of manganese treated in the same way.

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EXAMPLE 10.

A 25% aqueous solution of a polyethylene

polyethylene glycol 35000 in 25% aqueous solution	viscosity at 25°C	acid number referred to pure poly- ethylene glycol	content of formalde- hyde	total content of aldehydes cal. as acet- aldehyde in %	content of peroxides cal. as H ₂ O ₂ in %
before the heat test	400 cP	0.15	0.0004	0.016	0.0009
after 30 hrs. heat test without stabilizer	8 cP	56.5	0.029	0.27	0.006
after 30 hrs. heat test with 0.012% manganese powder	351 cP	1.3	0.004	0.009	0.0007

EXAMPLE 11.

15 0.1% of manganese powder referred to the weight of the polyethylene glycol having a grain size of <40 μ is added to a 1% aqueous solution of a polyethylene glycol having a molecular weight of about 500000 and heated to
20 80° C for 6 hours. Since polyethylene oxides

of such high molecular weight are increasingly submitted to decomposition under thermal conditions, in this case they are not treated with air or oxygen. In the following Table decomposition is shown by the decreasing of the vis-
cosity.

polyethylene oxide mol. weight 500 000 in 1% aqueous solution	viscosity of the solution at 25°C
before the heat test	165 cP
after 6 hours heating to 80°C without addition	8 cP
after 6 hours heating to 80°C with addition of 0.1% manganese powder referred to the weight of the polyethylene glycol	136 cP

EXAMPLE 12.

20% aqueous solutions of poly addition products of ethylene oxide to stearic acid or stearyl alcohol which have been stabilized with 0.01% of manganese powder having a grain size of

<40 μ are submitted to the heat test as described in Example 1 and simultaneously oxygen is passed through and then they are compared with the corresponding solutions without addition of manganese.

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tested product in 20% aqueous solution	acid number referred to the undilute product	content of formaldehyde %	total content of aldehydes, calculated as acetaldehyde %	content of peroxides cal. as H ₂ O ₂ %	result
Stearic acid +25 mols of ethylene-oxide before the heat test	0.6	0.0006	0.005	0.0006	water-clear solution
stearic acid +25 mols of ethylene-oxide without stabilizer after 30 hrs. heat test	106.0	0.0106	0.2	0.0074	milky white dispersion
Stearic acid +25 mols of ethylene-oxide with 0.01% manganese powder after 30 hrs. heat test	1.0	0.001	0.004	0.0011	almost clear solution
stearyl alcohol +25 mols of ethylene-oxide before the heat test	0.5	0.0007	0.007	0.004	water-clear solution
stearyl alcohol +25 mols of ethylene-oxide without stabilizer after 30 hours heat test	57.0	0.0134	0.1	0.007	milky white dispersion
stearyl alcohol +25 mols of ethylene-oxide with 0.01% of manganese powder after the heat test	1.0	0.002	0.004	0.0007	almost clear solution

EXAMPLE 13.

Polypropylene glycol having a medium molecular weight of 2000 is submitted to the heat test as described in Example 1 by simultaneously passing through oxygen in the

presence of 0.05% of manganese powder having a grain size of <40 μ and then compared with the corresponding non-stabilized product.

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polypropylene glycol 2000	viscosity at 25°C	acid number
before the heat test	325 cP	0.1
after 30 hours heat test without stabilizer	285 cP	12.4
after 30 hours heat test with addition of 0.05% of manganese powder	324 cP	0.6

EXAMPLE 14.

Pure polypropylene glycol having a medium molecular weight of 400 shows under the condition of a heat test mentioned in Example 1, phenomena of decomposition, which can be

read off from the acid number and the aldehyde content. The following survey shows that the addition of 0.05% of manganese powder (grain size $<40\mu$) largely prevents decomposition.

polyethylene glycol 400	acid number	aldehyde content, calculated as acetaldehyde	peroxide content calculated as H_2O_2
before the heat test	0.1	0.0073	0.0026
without stabilizer after 24 hours heat test	61.2	0.096	0.0550
with 0.05% of manganese powder after 24 hours of heat test	0.5	0.014	0.0054

WHAT WE CLAIM IS:—

1. A composition which comprises (a), a homo- or copolymer of an alkylene oxide, or a reaction product of an alkylene oxide with a substance containing active hydrogen, and (b) powdered manganese metal or a manganese-containing compound.

2. A composition as claimed in claim 1, which comprises from 0.0005 to 5% of component (b), based on the weight of component (a).

3. A composition as claimed in claim 1, which comprises from 0.005 to 0.5% of component (b), based on the weight of component (a).

4. A composition as claimed in any one of claims 1 to 3, wherein as component (b) there is used a manganese-(II) salt of an inorganic or organic acid.

5. A composition as claimed in claim 4, wherein the manganese-(II) salt is manganous chloride, manganous sulphate, manganous nitrate, manganous acetate or manganous resinate.

6. A composition as claimed in any one of claims 1 to 3, wherein as component (b) there is used manganese dioxide or potassium permanganate.

7. A composition as claimed in any one of claims 1 to 3, wherein as component (b) there is used powdered manganese metal having a particle size of less than 150μ .

8. A composition as claimed in claim 7, wherein the particle size of the manganese powder is less than 40μ .

9. A composition as claimed in any one of claims 1 to 8, wherein as component (a) there is used polyethylene glycol or polypropylene glycol.

10. A composition as claimed in any one of claims 1 to 8, wherein as component (a) there is used a copolymer of ethylene oxide and propylene oxide.

11. A composition as claimed in any one of claims 1 to 8, wherein the substance containing active hydrogen is an alcohol.

12. A composition as claimed in claim 11, wherein the alcohol is stearyl alcohol or oleyl alcohol.

13. A composition as claimed in any one of claims 1 to 8, wherein the substance containing active hydrogen is an acid.

14. A composition as claimed in claim 13, wherein the acid is stearic or oleic acid.

15. A composition as claimed in any one of claims 1 to 14, which is an aqueous solution of component (a).

16. A composition as claimed in claim 15, wherein the aqueous solution comprises a mineral acid.

17. A composition as claimed in claim 1, substantially as described in any one of the Examples herein.

18. A process for stabilizing a homo- or copolymer of an alkylene oxide, or a reaction product of an alkylene oxide with a substance containing active hydrogen, which comprises intimately mixing the homo- or copolymer or the reaction product with powdered manganese metal or a manganese-containing compound.

19. A process as claimed in claim 18, wherein the manganese metal or the manganese-containing compound is in the form of a solution or a dispersion.

20. A process as claimed in claim 18, the product of which is a composition as claimed in any one of claims 1 to 16.

21. A process as claimed in claim 18,
carried out substantially as described in any
one of the Examples herein.

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